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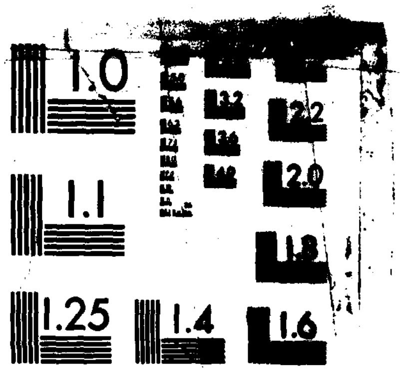

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MECHANISMS OF EXCITATION AND
IONIZATION PROCESSES IN SPUTTERING

FINAL REPORT

IGNATIUS S.T. TSONG

AUGUST 7, 1985

OFFICE OF NAVAL RESEARCH

CONTRACT NO. N00014-81-K-0844

ARIZONA STATE UNIVERSITY

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A. Research Goal

The primary objective of our research project is to understand the basic mechanisms governing the inelastic outershell processes, i.e. excitation, ionization and neutralization, in particle-solid interactions. To this end, we direct our investigation on the particles sputtered when a solid surface is subjected to energetic ion bombardment. An understanding of the basic mechanisms will allow better interpretation of experimental data obtained by surface analytical techniques which make use of sputtering such as secondary ion mass spectrometry (SIMS), Auger electron spectrometry (AES) and x-ray photoemission spectrometry (XPS). These techniques are commonly used to study microelectronics problems concerning interfairal properties and diffusion of dopants and impurities.

B. Summary of Work Accomplished

During the period from September 1, 1981 to February 28, 1985 supported by the Office of Naval Research, our efforts were largely focused on the study of molecules and clusters sputtered during ion bombardment. One of the most important questions in sputtering is whether molecules and clusters are ejected intact, i.e. the so-called "preformed" cluster, or they are formed by recombination of individual atoms as they leave the surface. In our experiments with excited molecular species ejected from surfaces of C, Si and Al bombarded by a N_2^+ ion beam, we measured the vibrational molecular spectra of CN, SiN and AlN. The observed excited molecular states CN $B^2\Sigma$, SiN $B^2\Sigma$ and AlN $A^3\Pi$, if formed from a recombination mechanism, will require at least one excited atom. The probability of such an occurence would be very low since excited state populations are much lower than ground state populations. We therefore conclude that the observed excited molecules are most likely preformed. This investigation was reported in our Progress Report #4 and Publication #6.

The preformed cluster mechanism is further confirmed by our theoretical study of the mass distribution of large molecular and cluster ions sputtered from biomolecules such as insulin and alkali halides such as CsI. Using our model of non-cascade sputtering, we successfully fit the mass spectra of bovine insulin ($\sim$5800 amu) obtained by the Uppsala and the Texas A & M groups and also the $(CsI)_nCs^+$ distribution obtained by Naval Research Lab and Manitoba groups. The non-cascade sputtering model basically considers the energy deposited by the incident primary particle to be partly channeled into vibrational excitation which in turn leads to particle ejection. Such a consideration is justified if we are dealing with an insulating or molecular solid in which fast energy conduction by free electrons is absent. And if the molecular or cluster ions are performed, i.e., no charge transfer has taken place during the ejection process, then one can apply the statistical RRK theory to explain and predict the observed mass distribution of the clusters. We have reported this work in Publications #8 and #9.

Our most recent work was on the sputtering of H_2O, CO_2 and N_2O ices at 78K by keV Ar^+, Ne^+, N^+ and He^+ ion bombardment and also 500eV electron bombardment. In measuring the total sputtering yield and the mass distribution of the sputtered ions, we find that there is a significant contribution from non-cascade process, i.e. processes other than nuclear stopping, to the observed yields. Very large clusters were observed, which could only be due to a "preformed" mechanism. This work was reported in our Progress Report #5 and Publication #10.

Our conclusion on completion of the research project is that large molecules and clusters in excited and ionized states ejected from insulating solids such as biomolecular films, alkali halides and ices are generally preformed, i.e., they are ejected intact. This is in contrast to the observation of ejection of cluster ions from metals, which can be attributed to a recombination mechanism as proposed by the Penn State group of Winograd and Garrison.

C. Technical Reports

1. Progress Report for period September 1, 1981 to April 30, 1982.
2. Progress Report for period May 1, 1982 to October 31, 1982.
3. Progress Report for period November 1, 1982 to May 31, 1983.
4. Progress Report for period June 1, 1983 to November 30, 1983.
5. Progress Report for period November 1, 1983 to May 31, 1984.

D. Publications

1. C.M. Loxton, I.S.T. Tsong and S.H. Lin. Commenet on "Molecule Formation During Sputtering by Two Body Associative Ionization with Diabatic Curve Crossing". Phys. Rev. Lett. 50, 1331 (1983).

2. A.R. Ziv, S.H. Lin, M. Skiff, B.P. Nigam, M. Szymonski, C.M. Loxton and I.S.T. Tsong. Theory of Inelastic Processes in Energetic Ion Impact on Solid Surfaces. J. Molecular Sci. 1, 55-70 (1983).

3. S.H. Lin, I.S.T. Tsong, A.R. Ziv, M. Szymonski and C.M. Loxton. Theoretical Studies of Non-Cascade Sputtering Processes. Phys. Scripta T6, 106-110 (1983).

4. C.M. Loxton, I.S.T. Tsong and H.W. Pickering. The Effect of Oxygen Adsorption on Cu-Ni Alloys During Irradiation Studied by SIMS, SIPS and ISS. Nucl. Instrum. Meth. 218, 340-346 (1983).

5. A.R. Ziv, B.V. King, S.H. Lin and I.S.T. Tsong. Kinetic Energy Distributions of Sputtered Particles in Non-Cascade Sputtering Processes. Nucl. Instrum. Meth. 218, 742-746 (1983).

6. C.M. Loxton, I.S.T. Tsong and D.A. Reed. Excitation of Molecules Formed by Ion Bombardment of Surfaces. Nucl. Instrum. Meth. B2, 465-469 (1984).

7. C.M. Loxton and I.S.T. Tsong. A Comparison of Secondary Ion and Photon Yields from Ion Bombarded Cu-Ni Alloys. Surface Sci. 139, 453-462 (1984).

8. B.V. King, A.R. Ziv, S.H. Lin and I.S.T. Tsong. Mass Distribution of Ejected Molecules and Clusters in Non-Cascade Sputtering Processes. J. Chem. Phys. 82, 3641-3645 (1985).

9. B.V. King, A.R. Ziv, S.H. Lin and I.S.T. Tsong. Interpretation of the Mass Distribution of Ejected $(CsI)_nCs^+$ Clusters by the Non-Cascade Sputtering Model. Surface Sci. (submitted).

10. J.W. Christiansen, D. Delli Carpini and I.S.T. Tsong. Sputtering of Ices by keV Ions. Nucl. Instrum. Meth. (submitted).

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